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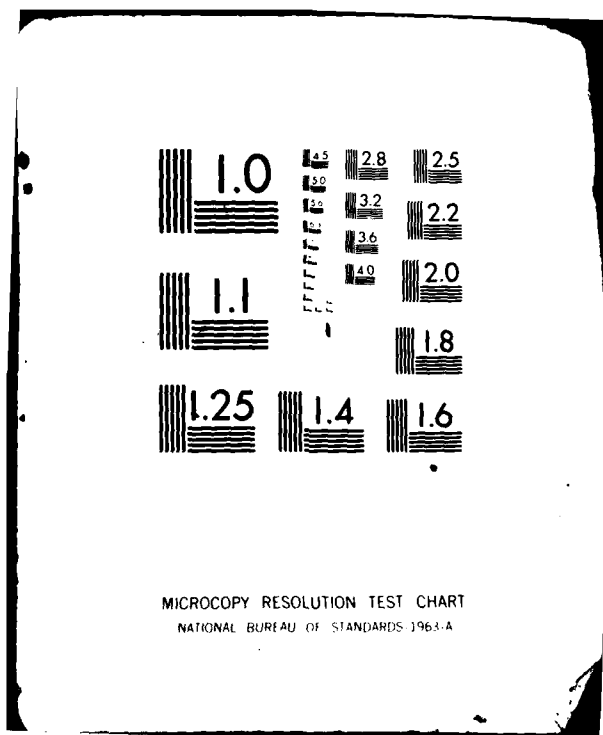
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PARTICLE BEAM CHEMISTRY STUDIES

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<p>Calculations were made of the chemistry in the channel left by a high intensity particle beam pulse. It was found that, because of the high temperature of the gas in the channel, the electron density was determined almost exclusively by electron-ion recombination. The role of nitrogen vibration in determining the temperature was also investigated.</p>		

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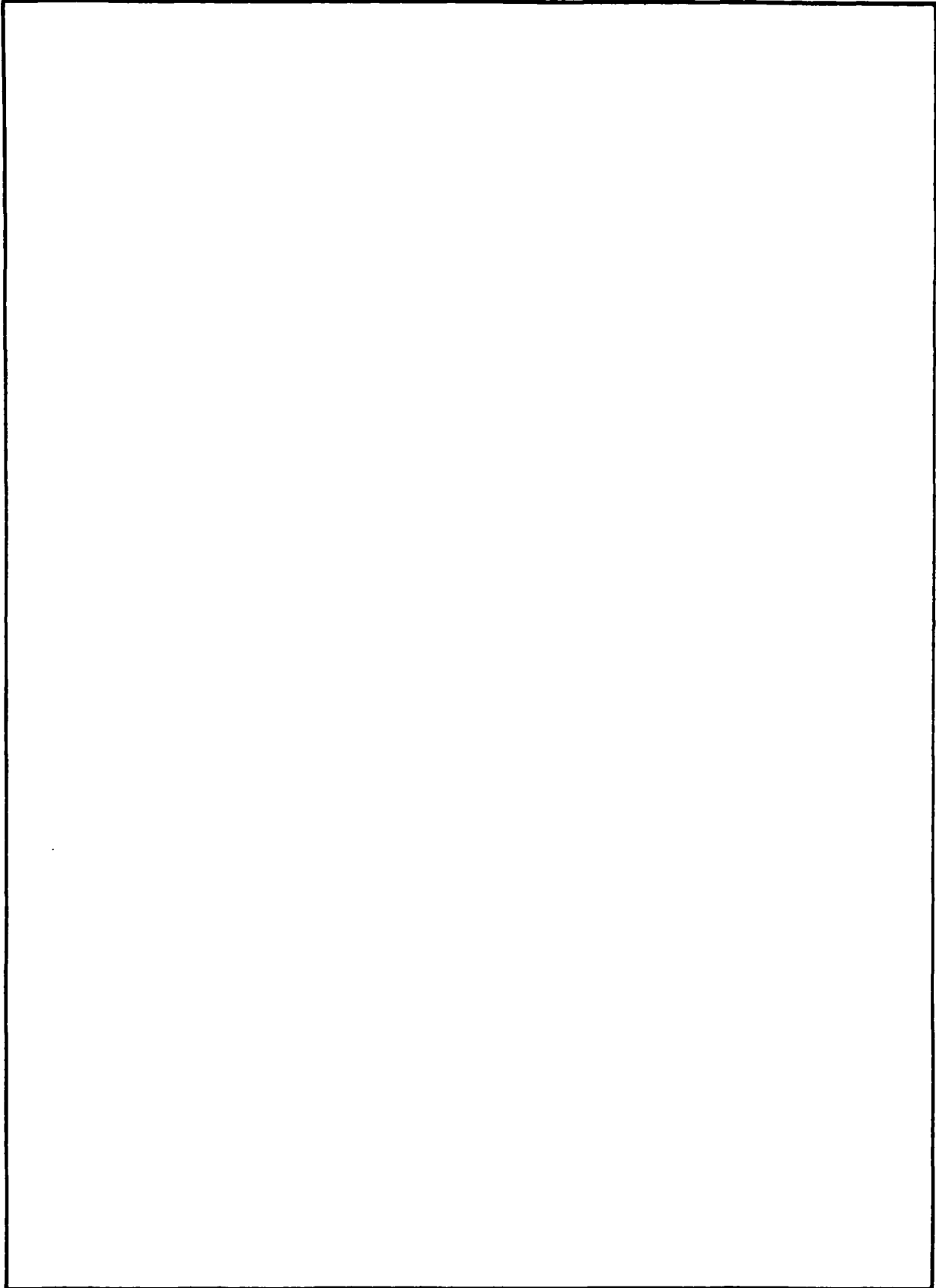
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## PARTICLE BEAM CHEMISTRY STUDIES

### INTRODUCTION

The successful propagation of a pulsed particle beam depends in part on the conditions in the air channel which is formed. In particular, the conductivity is a critical parameter, and this is determined primarily by the electron density. This report describes a short study which consisted of several chemistry calculations in the channel following an intense 2 mm radius beam and one calculation of the chemistry in the beam.

### CHANNEL INITIAL CONDITIONS

The temperatures and species concentrations at the end of the beam pulse were taken from a calculation made by SAI<sup>1</sup> for a 10 kA, 2 mm radius pulse. Although the pulse length was  $10^{-8}$  sec, the SAI calculation was carried out to  $2 \times 10^{-8}$  sec because of considerable ohmic heating which occurs after the high energy particle beam passes. The conditions at  $2 \times 10^{-8}$  sec were used as initial conditions in our calculation. The heavy particle temperature was 1810K, the electron temperature was 6380K and the nitrogen vibrational temperature was 5642K. The species concentrations are shown in Table 1.

The case chosen by SAI was a particularly intense beam and involves the deposition of a considerable amount of energy. The total energy deposited, including ohmic heating, is about  $3.5 \times 10^7$  ergs/cm<sup>3</sup>. About thirty percent of this energy is contained in N<sub>2</sub> vibration and thirty percent in dissociation and electronic excitation. This energy can be released by chemical reactions and excited state quenching during the



Table 1. Species concentrations at  $2 \times 10^{-8}$  sec.

E	=	3.06 (15)*
N <sub>2</sub>	=	1.97 (14)
N <sub>2</sub> (A)	=	7.60 (16)
O <sub>2</sub>	=	4.58 (18)
O <sub>2</sub> ( <sup>1</sup> Δ)	=	2.76 (17)
O <sub>3</sub>	=	2.03 (12)
N	=	4.17 (17)
N( <sup>2</sup> D)	=	5.17 (16)
O	=	1.02 (18)
O( <sup>1</sup> D)	=	6.01 (14)
NO	=	1.70 (17)
NO <sub>2</sub>	=	2.16 (14)
N <sup>+</sup>	=	2.36 (12)
N <sub>2</sub> <sup>+</sup>	=	8.60 (12)
N <sub>3</sub> <sup>+</sup>	=	1.04 (13)
N <sub>4</sub> <sup>+</sup>	=	4.82 (11)
NO <sup>+</sup>	=	2.34 (15)
O <sup>+</sup>	=	4.63 (10)
O <sub>2</sub> <sup>+</sup>	=	8.32 (14)
O <sup>-</sup>	=	4.45 (13)
O <sub>2</sub> <sup>-</sup>	=	8.82 (13)

\* Number in parenthesis is the power of ten.

evolution of the channel and can significantly impact the gas and electron temperatures.

## CHANNEL EXPANSION

The heating of the gas will cause an expansion of the channel. The initial heating of the gas to 1810K will cause an expansion (assuming adiabatic expansion, vibrational freezing and no further energy release) of about a factor of four and a concurrent cooling of the gas to about 1070K. This expansion takes several microseconds to occur but since we will be interested in tens of microseconds we have assumed the expansion takes place instantaneously. We therefore reduced all the species concentrations in Table 1 by a factor of four and used these values as our starting concentrations. In addition we made an estimate of the  $N_2O$  which would have formed during the energy deposition and added this to the initial conditions. This was done because  $N_2O$ , at these temperatures, is an excellent catcher of electrons. The value of  $N_2O$  we assumed after expansion was  $3.6 \times 10^{15} \text{ cm}^{-3}$ .

## CHEMISTRY CALCULATIONS

The chemical evolution of the channel was calculated using our SEACHEM code. This code integrates the coupled differential chemical equations and contains about 70 species and about 600 reactions. Since the chemical development will involve the release of energy stored in dissociation and vibration during the energy deposition period, the code keeps track of the energetics and allows for heating of the gas. It also allows for instantaneous adiabatic expansion of the gas as heating occurs. This density decrease amounted to factors of between 1.5 and 2 in our calculations, depending on the assumptions.

We will not list all the reactions in our code. As it turned out the high temperatures in the channel simplify the chemistry and only several reactions or processes are critical to the electron density.

When the ionization source turns off at  $10^{-8}$  seconds the dominant ion is  $O_2^+$ . Because of charge exchange reactions  $NO^+$  quickly becomes the dominant ion thereafter and by  $10^{-7}$  seconds is the only significant positive ion. The electron-ion recombination rate of  $NO^+$  is therefore an important rate constant. We have used a value of

$$k_1 = 4 \times 10^{-7} \left( \frac{300}{T_e} \right) \text{ cm}^3/\text{sec} \quad , \quad (1)$$

where  $T_e$  is the electron temperature. There is, however, considerable uncertainty in the dependence on  $T_e$ . Experimental values range from  $T_e^{-.4}$  to  $T_e^{-1}$ . For an electron temperature of about 0.5 eV or 6000K, this amounts to a difference in the rate constant of a factor of six. In a recombination dominated decaying plasma this amounts to a factor of six difference in the electron density.

Electron attachment is usually an important parameter in determining the electron density. The main attachment mechanism is usually three body attachment to  $O_2$ , i.e.,



The production of  $N_2O$ , however, in large amounts makes the following reaction potentially important,



Although this reaction is endothermic by about 0.2 eV, the presence of both hot electrons and vibrationally excited  $N_2O$  can make the rate constant for reaction 3 significant. Using the experimental data<sup>2</sup> and an electron temperature of several tenths of an eV we have obtained the rate constant

$$k_3 = 10^{-8} \left( \frac{T}{300} \right) e^{-3200/T} \text{ cm}^3/\text{sec}, \quad (4)$$

where T is the N<sub>2</sub>O temperature.

Detachment will also be important and the primary detachment mechanisms are



Reaction 4 is important because of the high temperatures involved in the channel.

Ion-ion recombination is usually dominated by the three-body process at sea level density and temperature. Because of the heating and expansion of the channel, however, the two-body process must be included. The effective two body rate constant used in our calculations was

$$k_9 = 6 \times 10^{-8} \left( \frac{300}{T} \right)^{0.5} + 10^{-25} \left( \frac{300}{T} \right)^{2.5} (M) \text{ cm}^3/\text{sec} \quad (9)$$

where T is the gas temperature and (M) is the total particle density in cm<sup>-3</sup>.

Finally, because of the energy stored in  $N_2$  vibration, the quenching of  $N_2$  vibration is an important parameter. Atomic oxygen seems to be the most efficient quencher of  $N_2$  vibration of all the species involved in our calculations. The measured rate constants between 1000K and 2000K can be fit by the expression<sup>3</sup>

$$k_{10} = 7.8 \times 10^{-15} \left( \frac{T}{300} \right)^{1.7} \text{ cm}^3/\text{sec} \quad (10)$$

A question which arises, however, is how efficient is the quenching by oxygen of the high vibrational states of  $N_2$ . If the quenching of the high vibrational states can also be given by the above expression then the  $N_2$  will lose a significant portion of its energy to translation in several tens of microseconds. If not it will take significantly longer.

## RESULTS

Four calculations were made. In two of the calculations the  $N_2$  vibration was assumed to be quenched slowly while in the other two the quenching was assumed to be rapid. Since the interchange of energy between electrons and nitrogen vibration is very rapid<sup>4</sup> we assumed that the electron temperature was equal to the  $N_2$  vibrational temperature. Thus the quenching of  $N_2$  controls the electron temperature. In each pair of calculations one calculation assumed no water vapor and the other assumed a 3 percent  $H_2O$  concentration. The  $H_2O$  was added to see if this made a significant difference in the electron density due to clustering of the positive ions and the formation of ions such as  $H_2O^+$  and  $H_3O^+$ . In addition, the inclusion of  $H_2O$  will yield species such as  $H$ ,  $OH$  and  $HO_2$ . These species can greatly effect the oxygen chemistry and therefore impact on the nitrogen vibrational quenching.

The electron densities for these four cases as a function of time (in microseconds) are given in Table 2. Cases A and B are for minimum  $N_2$  vibrational quenching. Case A is for no  $H_2O$  and case B is for 3 percent  $H_2O$ . Cases C and D are for maximum  $N_2$  vibrational quenching, with case C having no  $H_2O$  and case D, 3 percent  $H_2O$ .

Table 2. Electron densities.

t( $\mu$ sec)	Case A	Case B	Case C	Case D
10	2.1 (12)*	2.1 (12)	1.8 (12)	1.9 (12)
20	1.1 (12)	1.1 (12)	1.1 (12)	1.1 (12)
40	5.9 (11)	6.1 (12)	6.4 (11)	6.3 (11)
70	3.8 (11)	3.8 (11)	3.5 (11)	3.1 (11)
100	2.9 (11)	2.7 (11)	2.1 (11)	1.9 (11)

\* Number in parenthesis is power of ten.

We see that there is very little variation in the electron density among the different cases except at late times and then only by, at most, 50 percent. The addition of water vapor makes no significant difference in the type of ion which is present. Because of the high temperatures involved there is no clustering and  $NO^+$  remains the dominant ion. The addition of  $H_2O$  does make a significant difference in the recombination of atomic oxygen. The atomic oxygen disappears much more rapidly and this does decrease the  $N_2$  quenching. However, part of this decrease is offset by quenching by H and  $H_2$ . Also the more rapid recombination of oxygen adds to the heating of the gas and this offsets the reduced gas heating caused by the reduced  $N_2$  vibrational quenching. A more accurate

treatment of the interaction between the electron temperature and  $N_2$  vibration would probably make the differences between the water and no water cases larger but they would still not be significant.

The presence of  $N_2O$  does increase the attachment significantly at early times. At late times the high temperature has caused much of the  $N_2O$  to thermally dissociate. Also at late times the attachment, which is still large even without the  $N_2O$ , is offset by an equally large detachment caused in large part by the high temperature. Thus the electron density decay is electron-ion recombination dominated. The electron density is essentially determined by the dissociative recombination rate constant for  $NO^+$ .

#### CALCULATION WITHIN THE BEAM

The addition of water vapor in the previous calculations had no effect in changing the fact that  $NO^+$  was the dominant ion. However, the SAI calculation within the beam had no water vapor. The ions  $N_2^+$ ,  $O^+$  and  $N^+$  can charge exchange with water to form  $H_2O^+$  and this ion can react with water to form  $H_3O^+$ . We thought that the  $H_3O^+$ , which has a larger recombination rate than  $NO^+$ , formed within the beam might persist and contribute to the electron decay after the beam had passed. We therefore made a calculation within the beam. The ion-production rate specified by SAI was used (about  $4 \times 10^{17}$  ion-pairs/cm<sup>3</sup>/sec for  $10^{-8}$  seconds) as was their results for the gas and electron temperatures. The calculation showed that  $H_2O^+$  and  $H_3O^+$  were indeed significant within the beam but that these species decayed rapidly after the ionization source is turned off. By  $2 \times 10^{-8}$  seconds, the recombination rate was totally dominated by  $NO^+$  and  $O_2^+$ .

This calculation, however, did show significant differences in the concentrations of a number of other species at  $2 \times 10^{-8}$  seconds between

the SAI calculation and ours. Our atomic oxygen concentration is about half that obtained by SAI and this is due to H and OH chemistry. Our  $O_2(^1\Delta)$  concentration was only 40 percent the corresponding SAI value and this was probably due to our inclusion of quenching by electrons and perhaps a lower value of the  $O_2(^1\Delta)$  formed per initial ion-pair. These reduced values could impact on later detachment and  $N_2$  vibrational quenching, particularly in less intense, lower temperature beam cases.

Our  $N(^2D)$  concentration was about an order of magnitude smaller than the SAI value and this is undoubtedly due to the quenching of this state by  $H_2O$ . This quenching probably causes the species NH to be formed and this causes a whole new chemistry problem to be introduced since the chemistry of NH is largely unknown.

Our value of ozone was considerably larger than that in the SAI calculation. We obtained an  $O_3$  concentration of  $8.7 \times 10^{14} \text{ cm}^{-3}$ , which was about 400 times the SAI value. This large formation was due to the detachment reactions



This may be somewhat unrealistic since the reverse of these reactions were not included even though they are only about 0.6 eV endothermic. The rate constants, however, cannot be obtained from detailed balance since these reactions almost certainly involve  $O_3$  in vibrationally excited states. The dependence of the rate constants on the vibrational states is unknown and must be determined experimentally.

Finally the electron density we obtained at  $2 \times 10^{-8}$  seconds was  $8.3 \times 10^{14} \text{ cm}^{-3}$ , a factor of about four less than that in the SAI



calculation. This is due to the attachment to  $N_2O$  formed in our calculation. As we have already pointed out, however, this  $N_2O$  will be destroyed later by the high temperature.

## CONCLUSIONS

The main conclusion to be drawn from our calculations is that the chemistry of very intense beams is fairly simple due to the high temperatures involved. The electron concentration is primarily determined by the electron-ion recombination rate constant for  $NO^+$ . Unfortunately this rate constant is highly uncertain at high temperatures. We used a  $T_e^{-1}$  dependence. Use of a  $T_e^{-0.4}$  dependence would probably reduce the electron density to below  $10^{11}$  cm<sup>3</sup>/sec after 40 microseconds. This could have serious consequences for the stability of the second pulse trying to propagate in the channel at those times.<sup>5</sup> Another uncertainty resides in the electron temperature at microsecond times due to the uncertainty involved in the quenching of  $N_2$  vibration and its coupling to  $T_e$ . This should be examined more carefully.

There is considerable interest in less intense beams.<sup>5</sup> In these the heating will be much less severe and the chemistry in the channel will be more complicated. Cluster ions will be important and will persist out to late times. Attachment will be greater due to  $N_2O$  and probably  $O_3$ . This should be investigated in more detail. Detachment will be much less. The quenching of  $N_2$  vibration will be reduced and this will keep the electron temperature higher for a longer period of time. Previous calculations have indicated that it will take about three pulses to produce a highly ionized channel ( $n_e > 10^{11}$ ). However, in these calculations ohmic heating was not included,  $T_e$  was wrong,  $N_2$  vibration was not included, the expansion was not properly handled and the chemistry scheme was not as complete.

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